



10/06/00

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**  
**REQUEST FOR FILING NATIONAL PATENT APPLICATION**

Under 35 USC 111(a) and Rule 53(b)

A  
 PATENT APPLICATION

Asst. Commissioner of Patents  
 Washington, D.C. 20231

**WITH SIGNED DECLARATION**

**NONPROVISIONAL  
 NON REISSUE  
 NON PCT NAT PHASE**

jc825 U.S. PTO  
 09/680308

Sir:

Herewith is the PATENT APPLICATION of  
 Inventor(s): BOSMAN et al.

Title **PROCESS FOR THE HYDROGENATION OF PHENYL  
 ACETYLENE IN A STYRENE-CONTAINING MEDIUM WITH  
 THE AID OF A CATALYST**

Atty. Dkt.: PM 274361 | 9271US/CON/WO  
 M# Client Ref

including:

Date: October 6, 2000

1. Specification: 14 pages (only spec. and claims)
2.  Specification in non-English language
3. Declaration  Original  Facsimile/Copy  Abstract 1 page(s); 12 numbered claims
4.  Drawings: \_\_\_\_\_ sheet(s)  informal;  formal of size:  A4  11"
5.  See top first page re prior Provisional, National or International application(s). ("X" box only if info is there and do not complete corresponding item 5 or 6). (Prior M# \_\_\_\_\_ SN \_\_\_\_\_)
6. **AMEND the specification** please by inserting before the first line: -- This is a  Continuation-in-Part  
 Divisional  Continuation  Substitute Application (MPEP 201.09) of:  
 6(a)  National Appln. No. \_\_\_\_\_ / filed \_\_\_\_\_ . (M# \_\_\_\_\_)  
 6(b)  International Appln. No. PCT/NL99/00245 filed April 26, 1999
7.  **AMEND the specification** by inserting before the first line: -- This application claims the benefit of U.S. Provisional Application No. 60/ \_\_\_\_\_, filed \_\_\_\_\_ . --
8.  Attached is an assignment and cover sheet. Please return the recorded assignment to the undersigned.
9.  Prior application is assigned to \_\_\_\_\_

by Assignment recorded \_\_\_\_\_ Reel \_\_\_\_\_ Frame \_\_\_\_\_

10. **FOREIGN** priority is claimed under 35 USC 119(a)-(d)/365(b) based on filing in NL

11. \_\_\_\_\_ (country)

Application No.	Filing Date	Application No.	Filing Date
(1) 1009014	April 28, 1998	(2)	
(3)		(4)	
(5)		(6)	
(7)		(8)	
(9)		(10)	

12. 1 (No.) Certified copy (copies):  attached;  previously filed (date) \_\_\_\_\_  
 in U.S. Application No. \_\_\_\_\_ / filed on \_\_\_\_\_

13.  Attached: \_\_\_\_\_ (No.) Verified Statement(s) establishing "small entity" status under Rules 9 & 27.

14. DOMESTIC/INTERNATIONAL priority is claimed under 35 USC 119(e)/120/365(c) based on the following provisional, nonprovisional and/or PCT international application(s):

Application No.	Filing Date	Application No.	Filing Date
(1) PCT/NL99/00245	April 26, 1999	(4)	
(2)		(5)	
(3)		(6)	

15.  This application is being filed under Rule 53(b)(2) since an inventor is named in the enclosed Declaration who was not named in the prior application.

16.  Attached: Information Disclosure Statement; PTO-Form 1499; International Search Report; International Preliminary Examination Report and Preliminary Amendment

17.  Preliminary Amendment:

**THE FOLLOWING FILING FEE IS BASED ON CLAIMS AS FILED LESS ANY ABOVE CANCELLED**

	Large/Small Entity		Fee Code
18. Basic Filing Fee	\$710/\$355	\$710	101/201
19. Total Effective Claims 12 minus 20 = *0	x \$18/\$9 = + 0	+ 0	103/203
20. Independent Claims 1 minus 3 = *0	x \$80/\$40 = + 0	+ 0	102/202
	*If answer is zero or less, enter "0"		
21. If any proper multiple dependent claim (ignore improper) is present, add (Leave this line blank if this is a reissue application)	+ \$270/\$135	+ 0	104/204
22.	<b>TOTAL FILING FEE ENCLOSED =</b>	<b>\$710</b>	
23. If "non-English" box 2 is X'd, add Rule 17(k) processing fee	+ \$130	+ 0	139
24. If "assignment" box 8 is X'd, add recording fee	+ \$40	+ 40	581
25. <input type="checkbox"/> Attached is a Petition/Fee under Rule No.	+ \$130	+ 0	122
26.	<b>TOTAL FEE ENCLOSED =</b>	<b>\$750</b>	

Our Deposit Account No. 03-3975

Our Order No. 30268 0274361

C# M#

**CHARGE STATEMENT:** The Commissioner is hereby authorized to charge any fee specifically authorized hereafter, or any missing or insufficient fee(s) filed, or asserted to be filed, or which should have been filed herewith or concerning any paper filed hereafter, and which may be required under Rules 16-18 (missing or insufficient fee only) now or hereafter relative to this application and the resulting Official document under Rule 20, or credit any overpayment, to our Account/Order Nos. shown above for which purpose a duplicate copy of this sheet is attached.

This **CHARGE STATEMENT** does not authorize charge of the **issue fee** until/unless an issue fee transmittal form is filed.

**Pillsbury Madison & Sutro LLP**  
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NOTE: File in duplicate with 2 post card receipts (PAT-103) & attachments

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Continuation Patent Application of

BOSMAN et al.

Group Art Unit: unknown

Filed: October 6, 2000

Examiner: tba

FOR: PROCESS FOR THE HYDROGENATION OF PHENYL ACETYLENE IN A  
STYRENE-CONTAINING MEDIUM WITH THE AID OF A CATALYST

\* \* \* \* \*

October 6, 2000

**PRELIMINARY AMENDMENT**

Hon. Commissioner of Patents  
and Trademarks  
Washington, DC 20237

Sir:

Prior to substantive action on the merits, please amend the above-identified application as follows.

**IN THE CLAIMS:**

Please amend the claims as follows:

In claim 1, line 3, please delete "characterized in that" and substitute therefor  
-- wherein --.

In claim 2, line 1, please delete "characterized in that" and substitute therefor  
-- wherein --.

In claim 3, line 1, please delete "any one of claims 1-2" and substitute therefor  
-- claim 1 --; and on lines 1-2, please delete "characterized in that" and substitute therefor -- wherein --.

In claim 4, line 1, please delete “any one of claims 1-3” and substitute therefor -- claim 1 --; and on lines 1-2, please delete “characterized in that” and substitute therefor -- wherein --.

In claim 5, line 1, please delete “any one of claims 1-4” and substitute therefor -- claim 1 --; and on lines 1-2, please delete “characterized in that” and substitute therefor -- wherein --.

In claim 6, line 1, please delete “any one of claims 1-5” and substitute therefor -- claim 1 --; and on lines 1-2, please delete “characterized in that” and substitute therefor -- wherein --.

In claim 7, line 1, please delete “any one of claims 1-6” and substitute therefor -- claim 1 --; and on lines 1-2, please delete “characterized in that” and substitute therefor -- wherein --.

In claim 8, line 2, please delete “any one of claims 1-7” and substitute therefor -- claim 1 --; and on lines 1-2, please delete “characterized in that” and substitute therefor -- wherein --.

In claim 9, line 1, please delete “any one of claims 1-8” and substitute therefor -- claim 1 --; and on lines 1-2, please delete “characterized in that” and substitute therefor -- wherein --.

In claim 10, line 1, please delete “any one of claims 1-8” and substitute therefor -- claim 1 --; and on lines 1-2, please delete “characterized in that” and substitute therefor -- wherein --.

In claim 11, line 1, please delete “any one of claims 1-10” and substitute therefor -- claim 1 --; and on line 2, please delete “characterized in that” and substitute therefor -- wherein --.

In claim 12, line 1, please delete "any one of claims 1-10" and substitute therefor -- claim 1 --; and on line 2, please delete "characterized in that" and substitute therefor -- wherein --.

**REMARKS**

The amendments to the claims set forth above were made to remove the multiple dependencies and more closely comply with idiomatic English.

It is respectfully submitted that the present application is in condition for allowance and a Notice to that effect is courteously solicited. However, if any questions remain, the Examiner is encouraged to call the undersigned to expedite the prosecution of this application.

Respectfully submitted,  
PILLSBURY MADISON & SUTRO LLP

By



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# APPLICATION UNDER UNITED STATES PATENT LAWS

Atty. Dkt. No. PM 274361  
(M#)

Invention: **PROCESS FOR THE HYDROGENATION OF PHENYL ACETYLENE IN A STYRENE-CONTAINING MEDIUM WITH THE AID OF A CATALYST**

Inventor (s): **BOSMAN et al.**

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This is a:

- Provisional Application**
- Regular Utility Application**
- Continuing Application**
  - The contents of the parent are incorporated by reference
- PCT National Phase Application**
- Design Application**
- Reissue Application**
- Plant Application**
- Substitute Specification**  
Sub. Spec Filed \_\_\_\_\_  
in App. No. \_\_\_\_\_ / \_\_\_\_\_
- Marked up Specification re**  
Sub. Spec. filed \_\_\_\_\_  
in App. No. \_\_\_\_\_ / \_\_\_\_\_

## SPECIFICATION

M 10-05.00

AMENDED PAGE 1

PROCESS FOR THE HYDROGENATION OF PHENYL ACETYLENE  
IN A STYRENE-CONTAINING MEDIUM  
WITH THE AID OF A CATALYST

The invention relates to a process for the hydrogenation of phenyl acetylene in a styrene-containing medium with the aid of a catalyst and in the presence of hydrogen gas.

Styrene is often polymerized to polystyrene. In the styrene-containing medium which is used for this purpose, the phenyl acetylene causes undesirable side reactions during the polymerization, such as cross-linking of the polymer chains. It is therefore of importance to keep the phenyl acetylene content of the styrene-containing medium as low as possible.

The above-mentioned process for the hydrogenation of phenyl acetylene is known from JP-A-55,35368.

Said patent publication describes a process for the hydrogenation of phenyl acetylene in a mixture of styrene, phenyl acetylene and o-xylene with the aid of a palladium or nickel catalyst. In the specific Example hydrogenation is effected with the aid of a palladium catalyst on an alumina carrier.

A drawback of the use of a palladium catalyst is that a palladium catalyst, when used for the hydrogenation of phenyl acetylene in a styrene-containing medium with minor

M 10.05.00

AMENDED PAGE 2

amounts of impurities, rapidly loses its activity and thus has a short service life. This is a major drawback because it entails high catalyst regeneration costs. Further, regeneration of the catalyst means loss of production or the need to have a second reactor which is put on stream while the catalyst is being regenerated in the first reactor.

The aim of the invention is to provide a catalyst which does not present said drawback or only to a lesser extent.

The invention is characterized in that the catalyst is a supported nickel catalyst with a nickel content of 10-25 wt.%. Surprisingly it has appeared now that in a same styrene-containing medium a nickel catalyst is much less sensitive to impurities and thus has a much longer service life. This is all the more surprising in that a person skilled in the art, relying on his knowledge of catalysts, would not be likely to opt for a nickel catalyst instead of a palladium catalyst, since a nickel catalyst normally has a lower activity and selectivity than a palladium catalyst in the conversion of acetylenes to alkenes (see for instance J.A. Moulijn, P.W.N.M. van Leeuwen and R.A. van Santen, *Catalysis*, Elsevier, 1993, pp. 180-181).

Further advantages of application of a nickel catalyst are that the price of a nickel catalyst is lower than that of a palladium catalyst and that nickel catalysts are commercially available with a larger catalytically active surface area.

The nickel catalyst applied according to the invention is a supported nickel catalyst. Examples of suitable carrier materials are: silica,  $\alpha$ -,  $\theta$ - and  $\gamma$ -alumina, zeolites, carbon and oxidic carriers, such as for instance magnesium oxide, titanium oxide and zirconium oxide.

#### Mixtures of different carrier

5 materials can also be used. By preference,  $\theta$ - and  $\gamma$ -alumina, silica or carbon are used as carrier material. Particular preference is given to  $\theta$ - or  $\gamma$ -alumina as carrier material, because this is an inert carrier material with a large total surface area and a good  
10 poor volume distribution.

The nickel catalyst is synthesized for instance in the following way. The nickel is applied onto the carrier material by impregnating it with a solution of nickel salts. Water is commonly used as  
15 solvent. Then the impregnated carrier material is dried and subsequently calcinated at elevated temperature. The nickel oxide thus obtained is then activated on the carrier material through a treatment with hydrogen at elevated temperature. A high degree of dispersion of  
20 the nickel on the carrier results in a catalyst with a large catalytically active surface area. The higher the catalytically active surface area in the catalyst, the better the phenyl acetylene is hydrogenated.

For economical and technical reasons the  
25 nickel content of the nickel catalyst is kept as low as possible. A nickel catalyst with a higher nickel content is more expensive and when the nickel content becomes higher, the dispersion of the nickel in the catalyst is worse.

30 The nickel content of the catalyst is

normally 10-25 wt.%. Supported nickel catalysts are commercially available with for instance 10, 15 or 20 wt.% of nickel. The nickel content of the nickel catalyst preferably is 11 to 25 wt.%.

5 Most preferably the nickel content of the catalyst is more than 11 and less or equal to 20 wt.%.

Besides the nickel the catalyst can also contain minor amounts of other compounds which enhance the activity and selectivity of the catalyst. Examples 10 of such compounds are: chromium, gold, rhodium and ruthenium. The catalyst can also be modified with sulphur-containing compounds.

When its activity in the hydrogenation of phenyl acetylene has declined strongly, the nickel catalyst can be regenerated. Regeneration is effected 15 for instance by treating the contaminated catalyst with steam and air at a high temperature, for instance 300 to 350°C, followed by reduction with hydrogen at the same temperature. If the catalyst is lightly contaminated it can also be regenerated by merely 20 treating it with hydrogen at an elevated temperature of 100-300 °C.

When naphtha, gas condensates and LPG are cracked, cracked petrol is formed. This cracked petrol 25 mainly contains aliphatic and aromatic compounds with 6-9 carbon atoms, including 1-50 wt.% of styrene.

Fractionation of this cracked petrol yields a C8 fraction which contains 30-70 wt.% of styrene. When 30 this C8 fraction is extracted with a solvent and then distilled, a styrene-rich fraction is obtained which may contain more than 95 wt.% of styrene. The cracked

petrol, the C8 fraction as well as the styrene-rich fraction can be used as styrene-containing medium.

A styrene-containing medium can also be obtained by means of chemical synthesis. Alkylation of 5 benzene with ethene gives ethyl benzene, which through dehydrogenation can be converted into a styrene-containing medium which besides styrene can also contain ethyl benzene and phenyl acetylene.

The invention is not restricted, however, 10 to the hydrogenation of phenyl acetylene in the above-mentioned styrene-containing media. Styrene-containing media which have been obtained in another way can also be employed.

The styrene-containing media can comprise 15 up to 99.99 wt.% of styrene.

The styrene-containing medium preferably comprises more than 30 wt.% of styrene. By particular preference, the styrene-containing medium is a C8 hydrocarbon fraction which comprises more than 30 wt.% 20 of styrene.

In the hydrogenation of phenyl acetylene in the styrene-containing medium it is preferred for all phenyl acetylene in the styrene-containing medium to be converted to styrene or ethyl benzene, with a limited 25 amount of phenyl acetylene or styrene being hydrogenated to ethyl benzene.

The phenyl acetylene content of a styrene-containing medium is normally between 0.01 and 5 wt.% relative to the styrene present in the styrene-containing medium. During the hydrogenation process 30 according to the invention this content is reduced.

Said content is preferably reduced to less than 100 ~ ppm, by particular preference to less than 10 ppm in the styrene-containing medium.

The process for the hydrogenation of phenyl 5 acetylene in a styrene-containing medium is carried out in a reactor. The catalyst on a support is present in the reactor. The reactor is fed with the styrene-containing medium and hydrogen gas. The hydrogen gas can optionally have been diluted with another, inert 10 gas such as for instance nitrogen gas. The styrene-containing medium and the hydrogen gas can be mixed before being fed to the reactor.

The reactor can be operated as a two-phase or as a three-phase reactor.

15 If the reactor is operated as a two-phase reactor, then the hydrogen gas that is required for the hydrogenation of the phenyl acetylene is fully dissolved in the styrene-containing medium that is supplied to the reactor.

20 If the reactor is operated as a three-phase reactor the styrene-containing medium and the hydrogen gas are fed in at the bottom of the reactor and the product is obtained at the top of the reactor. It is also possible to supply the styrene-containing medium 25 and the hydrogen gas at the top of the reactor and to recover the product at the bottom of the reactor. The reactor can also be operated as a countercurrent reactor, with for instance the hydrogen gas being supplied at the bottom of the reactor and the styrene-containing medium at the top. The product is recovered 30 at the bottom of the reactor.

By preference the styrene-containing medium and the hydrogen gas are supplied at the bottom of the reactor, because there is less backmixing then and thus less styrene and phenyl acetylene react further to form 5 ethyl benzene.

The nickel catalyst is preferably present in a fixed bed in the reactor. The styrene-containing medium and the hydrogen gas are contacted with this fixed bed.

10 A fixed bed is a bed of solid catalyst parts that can have different shapes and can be, for instance, granules, pellets, extrudates, spheres, triloops and quadruloops. The fixed bed can also consist of a monolith or of minoliths.

15 The hydrogen pressure in the reactor is usually between 0 and  $300 \cdot 10^5$  Pa overpressure, preferably between 0 and  $50 \cdot 10^5$  Pa overpressure. Preferably a low pressure is applied, because then the reactor does not have to have a thick wall and is 20 therefore less costly.

The temperature is usually between 0 and 100 °C, preferably between 15 and 50 °C. If the temperature rises above 50 °C, polymerization of styrene occurs during the reaction.

25 The process according to the invention is preferably carried out with a hydrogen : phenyl acetate molar ratio  $\geq 1$ . By preference this molar ratio is between 1 and 10. For commercial-scale applications this molar ratio is preferably between 1 and 4. The 30 molar ratio is kept as low as possible in order to prevent conversion of styrene and phenyl acetylene into

ethyl benzene.

The average residence time of the styrene-containing medium in the reactor should not be too long, because then more styrene and/or phenyl acetylene react(s) further to ethyl benzene. The average residence time should not be too short either, because then the degree of conversion of phenyl acetylene to styrene is too low. A measure for the average residence time is the liquid hourly space velocity (LHSV).

The process according to the invention is usually carried out with a LHSV of between 0.1 and 100 per hour, preferably between 1 and 10 per hour.

The invention will now be elucidated by means of examples, without being restricted thereto.

15

Examples:

Example I

A reactor with a capacity of 1 m<sup>3</sup> was completely filled with a fixed bed consisting of a nickel catalyst on  $\theta$ -alumina. The catalyst contained 15 wt.% of nickel.

A C8 fraction comprising 50 wt.% of styrene, 8 wt.% of ethyl benzene and 0.8 wt.% of phenyl acetylene was supplied to the bottom of this reactor. Hydrogen gas was also supplied to the bottom of the reactor, the hydrogen gas : phenyl acetylene molar ratio being kept between 2 and 3. The LHSV was 4 h<sup>-1</sup> and the contact time was 15 minutes.

Further data of the reaction are presented in Table 1. The data were registered after different

on-stream times of the reactor. Measurements were carried out after 10, 100 and 220 days.

Comparative Example A

5           In the reactor of example I a hydrogenation reaction was carried out with a palladium catalyst instead of a nickel catalyst. A palladium catalyst on  $\gamma$ -alumina containing 0.2 wt.% of palladium was used. The other reaction conditions were the same as in  
10 example I. The service life of this catalyst was only 10 days. After 10 days the degree of conversion of phenyl acetylene had declined to the point where continuation of the hydrogenation reaction with this catalyst was not sensible any more.

15           Further data of the reaction are presented in Table 1.

Example II

20           In the reactor of Example I a hydrogenation reaction was carried out with the catalyst according to Example I after regeneration. The catalyst was regenerated by treating it with steam and air at 300 °C, followed by reduction with hydrogen at the same temperature.

25           The LHSV was 6  $\text{hr}^{-1}$  and the contact time was 10 minutes. The other reaction conditions were the same as in Example I.

30           Data after different on-stream times of the reactor were registered. Measurements were carried out after 10 and 100 days. Further data of the reaction are presented in Table 1.

Table 1

Example	t (days)	T <sub>in</sub> (°C)	T <sub>out</sub> (°C)	ΔT (°C)	X <sub>ph</sub> (%)	P <sub>h<sub>out</sub></sub> (ppm)
I	0	28	46	18	99.8	< 10
	10	28	46	18	99.8	< 10
	100	34	49	15	99.7	< 20
	220	39	53	14	99.6	< 30
A	0	20	40	20	99.9	< 10
	10	22	28	6	75.0	200 - 300
II	0	30	45	15	99.8	< 10
	10	30	45	15	99.8	< 10
	100	35	48	13	98.9	100

Explanation of the symbols:

5 t = time

T<sub>in</sub> = inlet temperature of the styrene-containing medium

T<sub>out</sub> = outlet temperature of the styrene-containing medium

10 ΔT = difference between the inlet and the outlet temperature

X<sub>ph</sub> = conversion of phenyl acetylene

P<sub>h<sub>out</sub></sub> = the phenyl acetylene content of the styrene-containing medium leaving the

15 reactor

Example III

A 95 ml reactor was filled with a fixed bed of 70 ml consisting of nickel catalyst on θ-alumina.

The catalyst contained 15 wt.% of nickel.

At the bottom of the reactor a C8 fraction was supplied, containing 50 wt.% of styrene, 8 wt.% of ethyl benzene and 0.8 wt.% of phenyl acetylene. The LHSV was 5 hr<sup>-1</sup>, the hydrogen pressure was 3 bar and the inlet temperature was 40 °C. Hydrogen gas was also supplied via the reactor bottom, with a varying hydrogen gas / phenyl acetylene molar ratio, as indicated in Table 2. From Table 2 it appears that at a higher hydrogen gas / phenyl acetylene molar ratio the conversion of phenyl acetylene increases. Some styrene is reacted further to ethyl benzene, however, causing the net styrene yield to decrease a little again.

15    Example IV

Example III was repeated, using a nickel catalyst on  $\theta$ -alumina containing 20 wt.% of nickel instead of 15 wt.%. The inlet temperature was 30 °C. The other reaction conditions were the same as in Example III. Hydrogen gas was supplied via the reactor bottom, with a varying hydrogen gas / phenyl acetylene molar ratio, as indicated in Table 2. From Table 2 it appears that at a higher nickel content of the catalyst the same degree of conversion of phenyl acetylene is obtained with a lower hydrogen / phenyl acetylene molar ratio. From this example it also appears that when the hydrogen / phenyl acetylene molar ratio is too high, styrene is also hydrogenated.

Table 2

Example	$H_2 / ph$ (mol/mol)	$X_{ph}$ (%)	S (%)
III	3	98.5	0.3
	5	99.9	0.1
	7	99.9	0.1
IV	1	91	0.4
	2	99.9	0.2
	3	99.9	-0.2

Explanation of the symbols:

5     $X_{ph}$  = conversion of phenyl acetylene  
H<sub>2</sub>/ph = molar ratio hydrogen / phenyl acetylene  
S = net styrene yield in the medium leaving the reactor relative to the medium supplied to the reactor

10

Example V

Example III was repeated at elevated temperature and with a LHSV of 5.5 hr<sup>-1</sup>. At a temperature of 70 °C the conversion of phenyl acetylene 15 was 94% at the start of the experiment. After 5 days the conversion of phenyl acetylene had declined to 30%.

At a temperature of 90 °C the conversion of phenyl acetylene was 100% at the start of the experiment. After 4 days the conversion of phenyl acetylene had declined to 40%. 20

The strong decline of the conversion of phenyl acetylene is due to formation of polystyrene on the catalyst surface at higher temperatures, resulting in loss of catalyst activity.

Enclosure 1.1

10.08.00

AMENDED SET OF CLAIMS

1. Process for the hydrogenation of phenyl acetylene in a styrene-containing medium with the aid of a catalyst and in the presence of hydrogen gas, characterized in that the catalyst is a nickel catalyst with a nickel content of 10-25 wt.% supported on a carrier material and in that the hydrogen gas / phenyl acetylene molar ratio is 1-10.
2. Process according to claim 1, characterized in that the nickel content of the catalyst is 11-25 wt.%.
3. Process according to any one of claims 1-2, characterized in that the carrier material is  $\theta$ - or  $\gamma$ -alumina.
4. Process according to any one of claims 1-3, characterized in that the catalyst is a fixed-bed catalyst with which the styrene-containing medium and the hydrogen gas are contacted.
5. Process according to any one of claims 1-4, characterized in that the styrene-containing medium and the hydrogen gas are supplied at the bottom of the reactor.
6. Process according to any one of claims 1-5, characterized in that the hydrogen gas / phenyl acetylene molar ratio is 1-4.
7. Process according to any one of claims 1-6, characterized in that the phenyl acetylene content of the styrene-containing medium is 0.01-5 wt.%.
8. Process according to any one of claims 1-7, characterized in that the temperature is between 15 and 50 °C.
9. Process according to any one of claims 1-8, characterized in that the LHSV is between 0.1 and 100 per hour.
10. Process according to any one of claims 1-8, characterized in that the LHSV is between 1 and 10 per hour.
11. Process according to any one of claims 1-10, characterized in that the styrene-containing medium contains  $\geq$  30 wt.% of styrene.

10-05-2000

- 14 -

NL 009900245

9271WO

Enclosure 1.2

**M 10.06.00**

12. Process according to any one of claims 1-10,  
characterized in that the styrene-containing medium is a  
C8 hydrocarbon fraction containing  $\geq$  30 wt.% of styrene.

A B S T R A C T

The invention relates to a process for the  
5 hydrogenation of phenyl acetylene in a styrene-  
containing medium with the aid of a supported nickel  
catalyst with a nickel content of 10-25 wt.%. This  
process is by preference used for the hydrogenation of  
phenyl acetylene in a styrene-containing medium which  
10 contains more than 30 wt.% of styrene.

As a below named inventor, I hereby declare that my residence, post office address and citizenship are as stated below next to my name, and I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the INVENTION ENTITLED:

PROCESS FOR THE HYDROGENATION OF PHENYL ACETYLENE IN A STYRENE-CONTAINING MEDIUM WITH THE AID OF A CATALYST

the specification of which (CHECK applicable BOX(ES))

→  is attached hereto.

X

→  was filed on \_\_\_\_\_

as U.S. Application No. \_\_\_\_\_

/

BOX(ES)

was filed as PCT International Application No. PCT/

NL99/00245

on \_\_\_\_\_

April 26, 1999

→  and (if applicable to U.S. or PCT application) was amended on \_\_\_\_\_

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above. I acknowledge the duty to disclose all information known to me to be material to patentability as defined in 37 C.F.R. 1.56. I hereby claim foreign priority benefits under 35 U.S.C. 119/365 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate filed by me or my assignee disclosing the subject matter claimed in this application and having a filing date (1) before that of the application on which priority is claimed, or (2) if no priority claimed, before the filing date of this application:

PRIOR FOREIGN APPLICATION(S)

<u>Number</u>	<u>Country</u>	<u>Day/MONTH/Year Filed</u>	<u>Date first Laid-open or Published</u>	<u>Date Patented or Granted</u>	<u>Priority Claimed</u>
1009014	the Netherlands	28 APRIL 1998		Oct. 29, 1999	<input type="checkbox"/>
					<input type="checkbox"/>
					<input type="checkbox"/>

I hereby claim domestic priority benefit under 35 U.S.C. 119/120/365 of the indicated United States applications listed below and PCT international applications listed above or below and, if this is a continuation-in-part (CIP) application, insofar as the subject matter disclosed and claimed in this application is in addition to that disclosed in such prior applications, I acknowledge the duty to disclose all information known to me to be material to patentability as defined in 37 C.F.R. 1.56 which became available between the filing date of each such prior application and the national or PCT international filing date of this application:

PRIOR U.S. PROVISIONAL, NONPROVISIONAL AND/OR PCT APPLICATION(S)

<u>Application No. (series code/serial no.)</u>	<u>Day/MONTH/Year Filed</u>	<u>Status</u>	<u>Priority Claimed</u>
PCT/NL99/00245	26 APRIL 1999	<u>pending, abandoned, patented</u>	<input type="checkbox"/>
		pending	<input type="checkbox"/>
			<input type="checkbox"/>
			<input type="checkbox"/>

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

And I hereby appoint Pillsbury Madison & Sutro LLP, Intellectual Property Group, 1100 New York Avenue, N.W., Ninth Floor, East Tower, Washington, D.C. 20005-3918, telephone number (202) 861-3000 (to whom all communications are to be directed), and the below-named persons (of the same address) individually and collectively my attorneys to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith and with the resulting patent, and I hereby authorize them to delete names/numbers below of persons no longer with their firm and to act and rely on instructions from and communicate directly with the person/assignee/attorney/firm/ organization who/which first sends/sent this case to them and by whom/which I hereby declare that I have consented after full disclosure to be represented unless/until I instruct the above Firm and/or a below attorney in writing to the contrary.

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